Nano-Silica Modified Recycled Aggregate Concrete: Mechanical Properties and Microstructure

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Abstract: Sustainability of concrete could be enhanced by incorporating recycled aggregate in the production of concrete. The aim of this study is to assess the effect of using nano-silica (nanoparticles of SiO_2) to improve the performance of recycled aggregate concrete (RAC) containing recycled aggregate (RA) derived from processing construction and demolition waste of concrete buildings. In this study, compressive strength of concrete and the microstructure of RA and RAC with and without nanoparticles of SiO_2 were examined. Nine mixes: one control mix with natural aggregate without nanoparticles, two mixes with RA at 50% and 100% contents (without nano-silica) and three mixes for each content of RA with nano-silica content of 0.4, 0.8 and 1.2% (by mass of cement) were investigated. The results showed that nanoparticles of silica can improve the compressive strength and modify the microstructure of RAC.

Keywords: **Nanoparticles SiO_2, Compressive Strength, Recycled Aggregate, SEM**

1. Introduction

The world is currently facing a global sustainability and environmental issue due to the increasing generation of massive quantities of waste by construction and demolition activities. More than 500 million of tons of construction and demolition waste (CDW) are generated worldwide annually (Younis & Pilakoutas, 2013). Such huge quantity of CDW requires substantial areas of landfill to dispose, causing serious environmental issues. Also, depletion of natural resources of natural aggregates used in the production of concrete contributes to other environmental and sustainability concerns. These environmental issues and concerns have led the researchers worldwide to examine methods to mitigate the impact of these issues on the environment. One way is recycling the CDW and reusing them as aggregate and as an alternative to the natural aggregate in concrete. The utilization of recycled aggregate (RA) in concrete can save the environment through conserving the scarce landfill areas and reduce the consumption rate of the natural resources of aggregates (Younis & Pilakoutas, 2013; Tam, 2008). The use of the RA generated from processing CDW of concrete elements in new concrete has been investigated for decades. Nonetheless, the properties of the recycled aggregate concrete (RAC), as reported by the researchers, are inferior to that of the natural aggregate concrete (NAC). RAC exhibits low compressive, tensile and flexural strength as well as high water absorption, porosity and shrinkage (Younis & Pilakoutas, 2013; Tam, Gao, & Tam, 2005). Many researchers have attributed this performance of RAC to the heterogeneous nature of the RA caused by the attached mortar. The attached mortar is characterized by high porosity, microcracks and flaws which make the RA particle weak and having loose microstructure. Weak interfacial transition zone (ITZ) between the recycled concrete aggregate (RCA) and the cement matrix has also been identified (Younis & Pilakoutas, 2013; Tam, Gao, & Tam, 2005; Katz, 2004). Therefore, researchers have tried various approaches to improve the properties of RAC (Katz, 2004). Some have examined the utilization of reactive, fine and ultrafine cementitious materials such as fly ash, ground granulated blast furnace slag and silica fume (Katz, 2004; Berndt, 2009). It is reported that these materials help to improve the performance of RAC due to the pozzolanic reaction and the filling ability resulting in a dense microstructure and strong ITZ (Tam, Gao, & Tam, 2005). However, the use of these materials in RAC does not always lead to a performance comparable to that of NAC. Hence, the need for investigating other materials is vital; nanomaterial, especially Nano SiO2 (Nano silica) is a promising material in this regard. Owing to its very small size (in the range of nanometers) and the pozzolanic reaction, Nano silica particles are very effective in enhancing the performance of concrete (Sanchez & Sobolev, 2010). Nanomaterial, particularly nano SiO_2, has the ability to improve the strength and the durability of concrete through accelerating the hydration reaction and filling the micropores in the cement paste structure. Studies (e.g. Sanchez & Sobolev, 2010; Said et al., 2012; Shaikh, Odoh, & Than, 2014) have shown that the addition of nano-silica results in increased compressive, tensile and flexural strength of normal concrete. Studies on the use of nano-silica to upgrade the performance of RAC are rare. Thus, this study aims at investigating the effect of using nano SiO_2 to improve the mechanical properties and modify the microstructure of RAC.

2. Materials And Experimental Programme

2.1 Materials

Cement

Portland Cement (CEM I type) was used in this study meeting the requirements of Iraqi specifications (IQS No. 45/1984). The cement is manufactured by Mas company located in Sulymania –Iraq.

Aggregates

Two types of coarse aggregates were used, natural and recycled. The natural gravel was rounded river aggregate with a maximum size of 20 mm, water absorption of 0.8% and specific gravity of 2.6. The coarse RA was recycled concrete aggregate produced by crushing old concrete portions generated by demolishing old concrete buildings. The water absorption and the specific gravity of the RA were 3.3% and 2.45, respectively. The fine aggregate used in this study was sand with a maximum size of 5 mm.

Nano Materials

The materials phase used in this study was nano-silica supplied by HWNANO company in China. The nano SiO_2 was in the form of powder with purity of 99% SiO_2 (see Figure.1). The average particle size of the nan-silica was between 20- 30 nm with surface area of 125 m^2/g. The X-ray diffraction (XRD) analysis of a powder sample of the SiO_2 nanoparticles is shown in Figure 1.

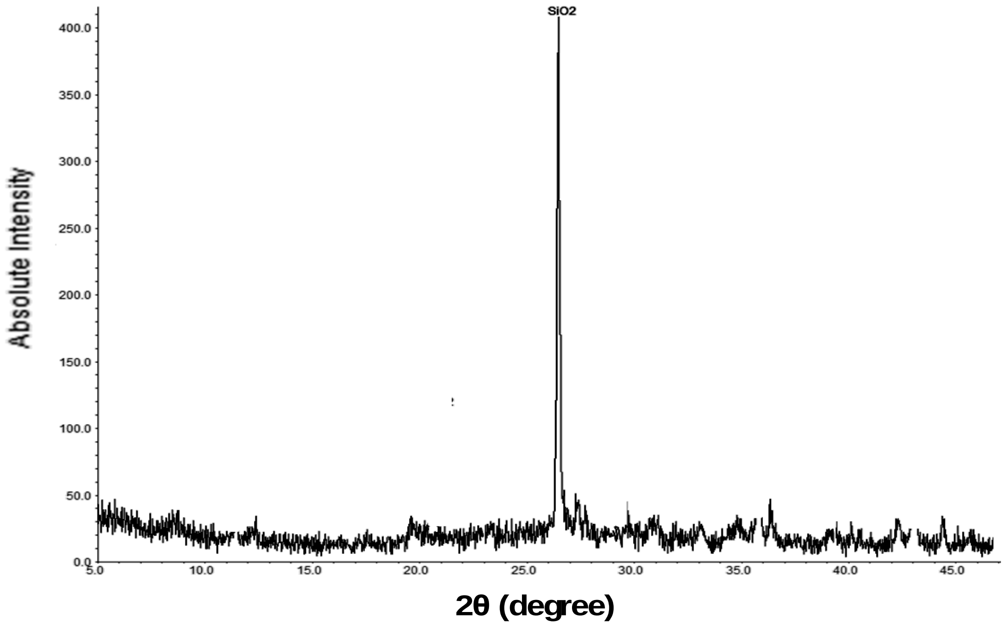


Figure 1: XRD analysis of SiO2 Nanoparticles used in the study

Superplastisizers

Superplasticizers were used to ensure a uniform dispersion of the Nanoparticles as explained in section 2.3. The Superplasticizer was an aqueous solution containing polycarboxylate ether (PCE) polymers.

2.2 Variables and Mix Proportions

In total, nine different concrete mixes were prepared. The code of mixes and the variables of the study are shown in Table 1. The mixes are divided into three groups according to the type of coarse aggregate, content of coarse recycled aggregate and content of nano materials (Nano SiO_2). The study includes: one control mix made with natural coarse aggregate NCA (R0), two mixes made with recycled coarse aggregate RCA in which the NA is replaced by RA at contents of 50% and 100% and six mixes (three for each RA content) made with three contents (0.4%, 0.8%and 1.2% by cement mass) of Nano SiO2. All mixes had the same quantity of cement (400 kg/m^3), water (192 kg/m^3) and fine aggregate (719 kg/m^3), whilst the quantity of coarse aggregate used was 1125 kg/m^3. All mixes made with the same water/cement (w/c) ratio (0.48)

Table 1: Code of mixes and variables of study

Mix no.	Mix code	Type of coarse aggregate	Nano SiO_2 content %[a]	Coarse recycled aggregate content (%)
1	R0	Natural	0	0
2	R50	Natural+Recycled	0	50
3	R100	Recycled	0	100
4	R50N0.4	Natural+Recycled	0.4	50
5	R50N0.8	Natural+Recycled	0.8	50
6	R50N1.2	Natural+Recycled	1.2	50
7	R100N0.4	Recycled	0.4	100
8	R100N0.8	Recycled	0.8	100
9	R100N1.2	Recycled	1.2	100

[a] By cement mass.

2.3 Mixing, Specimens Preparation and Curing

A pan mixer with capacity of 0.1 m^3 was used to mix the ingredients and prepare all concrete mixtures. For the mixes containing Nano SiO_2, the nano silica powder was added to a one liter of water and an amount of super plasticizer (0.5% by mass of cement) to form an aqueous solution. The solution was mixed using a high speed blender to ensure a uniform distribution of the Nano particles and avoiding the agglomeration of these particles. The procedure of mixing the ingredients of the concrete was as follows: firstly, the coarse aggregate was added and mixed with the aqueous solution of Nano SiO_2 and part of the mixing water for 1 minute. Then, the ingredients were left in the mixer pan for 10 minutes to allow the recycled coarse aggregate to absorb the Nano particles and form a coat layer on their surfaces. Secondly, the fine aggregate, cement and the rest of the mixing water were added and mixed for 3 minutes. After the completion of mixing the ingredients, for each mix: three (100 mm) cubes were cast. The concrete was mixed using a pan mixer and compacted using a vibrating table. After casting, the specimens were then covered by plastic sheets and allowed to cure for 24 hours before being demoulded. Then, the specimens were kept in water tanks for 27 days for further curing.

2.4 Tests

Compressive Strength Test

The compressive strength was obtained at age of 28 days using the 100 mm cubes according to BS EN 12390-3 (BS EN 12390-3).

Microstructure Observations

Individual particles of recycled coarse aggregate were investigated using a digital microscope. Scanning electron microscopy (SEM) was also used to assess the microstructure of the RA particles and concrete samples with and without Nanoparticles of SiO_2.

3. Results and Discussion

3.1 Compressive Strength

The results of the compressive strength at the age of 28 days for all mixes are presented in Table 2. The result of each mix is the average of three specimens. The table also shows the normalized strength (to that of the plain concrete, mix R0).

Table 2: Results of compressive strength test

Mix	Compressive strength	
	Strength in (MPa)	Normalized Strength
R0	40.6	1.00
R50	34.1	0.84
R100	30.2	0.74
R50 N0.4	37.5	0.92
R50 N0.8	40.3	0.99
R50N1.2	41.0	1.00
R100N0.4	32.0	0.79
R100N0.8	34.2	0.84
R100N1.2	35.1	0.86

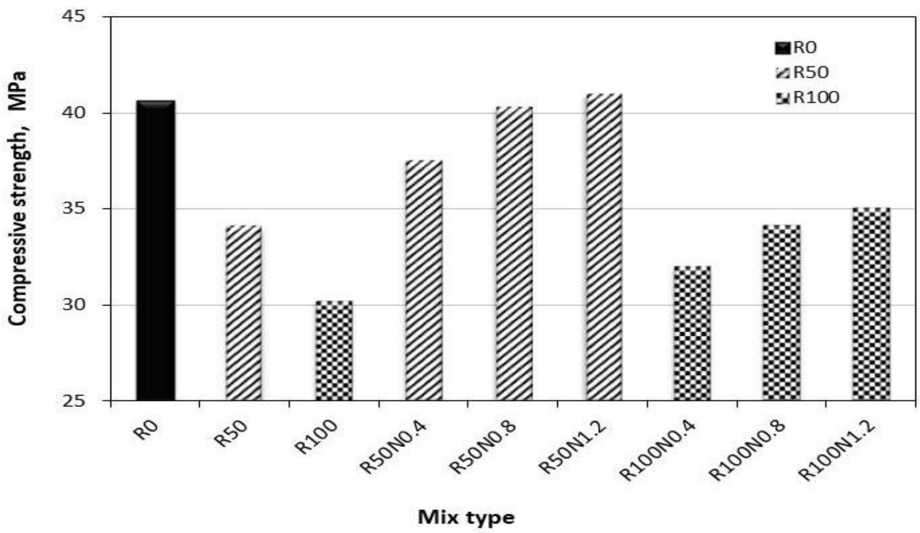


Figure 2: Compressive strength of all mixes

As expected, the compressive strength of the RAC mixes (R50, R100) is lower than that of the control mix (R0), as can be seen in Table 2 and Figure 2. This is mainly due to the heterogeneous nature of the recycled aggregate which is characterized by a weak and cracked surfaces resulted from the crushing process of demolished concrete (Tam, Gao, & Tam, 2005; Katz, 2004). The compressive strength of RAC decreased by 16% and 26% when the natural aggregate was replaced by 50% and 100%, respectively.

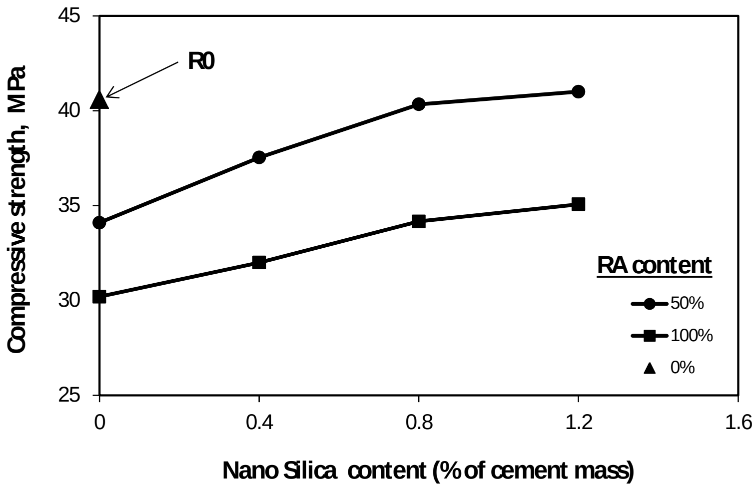


Figure 3: Effect of Nano silica particles content on the increase of compressive strength

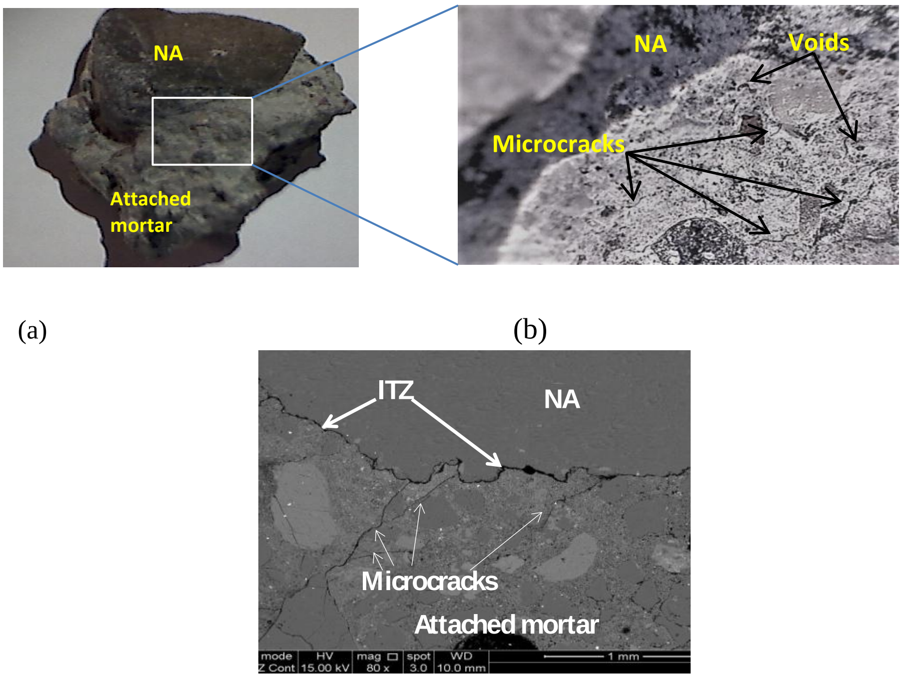
Figure 3 also shows that the addition of Nano SiO_2 enhanced the compressive strength of the mixes containing recycled aggregate. It is clear that the content of Nano particles affects the degree of strength enhancement. It can be seen that the compressive strength increases with the increase of content of Nano silica regardless of the recycled aggregate content. For example, when the Nano silica is added at contents of 0.4%, 0.8% and 1.2%, the compressive strength increases by 10%, 18% and 20% for mixes containing 50% RA and by 6%, 13%and 16% for mixes made with 100% RA. The results also show that the increase in strength for mixes containing 50% RA is higher than those with 100% RA at all Nano silica contents. It seems that the addition of Nano silica at contents up to 0.8% (by mass of cement) to the RAC mixes is very beneficial and can result in comparable strength to that of the control mix R0 (mix with natural coarse aggregate). The mechanisms leading to this improvement is explained in the next section where the effect of adding the nanoparticles on the microstructure of the RAC is discussed.

3.2 Microscopic and Scanning Electron Microscopy (Sem) Observations

3.2.1 Recycled Aggregate Specimens (Particles)

The surface of some individual particles of the recycled aggregate with and without Nano silica was observed by a microscope to support and justify the gained enhancement in the compressive strength for the mixes made with recycled coarse aggregate. Also, SEM images for recycled aggregate concrete specimens were taken and used for the same purpose. Figure 4a shows an image taken by a microscope with a magnification of 20x for the surface of a recycled aggregate particle.

Microcracks and voids can be identified on the surface of the RA particle as can be seen in Figure 4b. These microcracks and voids are the main cause of the weakness of the RA which in turn results in low strength concrete (Tam, Gao, & Tam, 2005; Katz, 2004). A weak and cracked zone between the natural aggregate (NA) and the attached mortar (ITZ) can be observed. These microcracks and weak ITZ are usually caused by the process of the crushing of the demolished concrete (Tam, Gao, & Tam, 2005). Similarly, microcracks and weak ITZ can be identified in Figure 4c which shows an SEM image for the surface of a particle of RA.



(a) (b)

(c)

Figure 4: a- RA particle at 20x magnification b- RA surface at 200x magnification
c- SEM image of the surface of a recycle aggregate particle

To assess the effect of adding the Nano silica as a coat layer around the RA particles on their surfaces, microscopic images were taken for individual particles as can be seen in Figure 5. The effect of the nano silica on the surface of the RA particles is clear. The Nano silica modifies the surface by sealing all the microcracks and filling all the voids. This modification can partly explain the enhancement in the strength gained by adding the Nano silica to the mixes with RA.

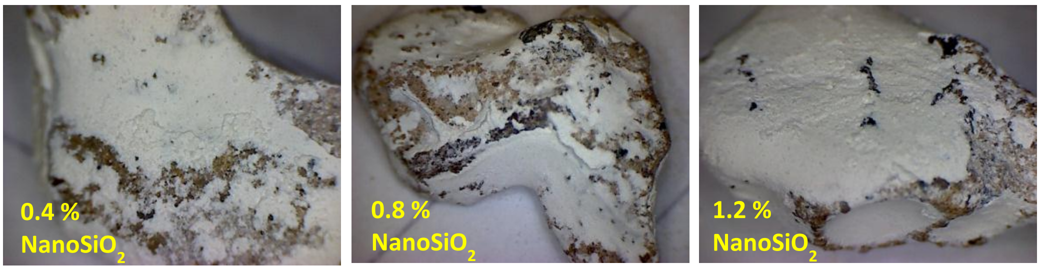


Figure 5: Effect of Nano silica content on the coating layer of RA particles

3.2.2 Concrete Specimens

The effect of using SiO_2 nanoparticles on the microstructure of RAC was also evaluated through observing concrete specimens with and without Nano SiO_2. The SEM images for the concrete specimens were taken and presented in Figure 6 and Figure7. Figure 6 shows SEM image for the RAC without SiO2 nanoparticles. The image clearly shows the fact that there are different ITZs in RAC: ITZ1 which exist between the NA particle and the old attached mortar ; ITZ2 which lays between the NA particles and the new cement matrix (mortar) and ITZ3 which develops between the new and the old cement mortars. The existence of these ITZs with different microstructures increases the heterogeneous nature of RAC and hence results in a weak concrete in terms of both strength and durability aspects. Also, the image shows that ITZ3 is like the other two ITZs is porous and can be clearly identified by the very fine black line. The high porosity of this ITZ is due to the high concentration of $Ca(OH)_2$ resulted from the hydration of cement which is known for its high porosity.

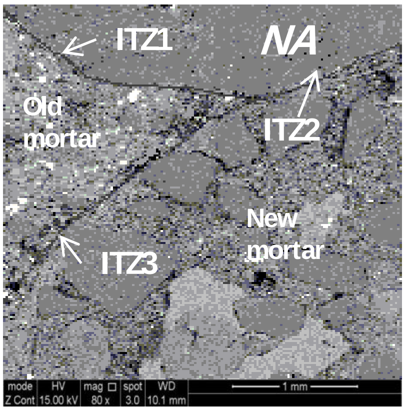
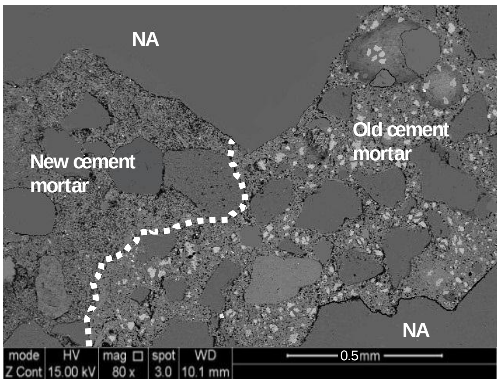
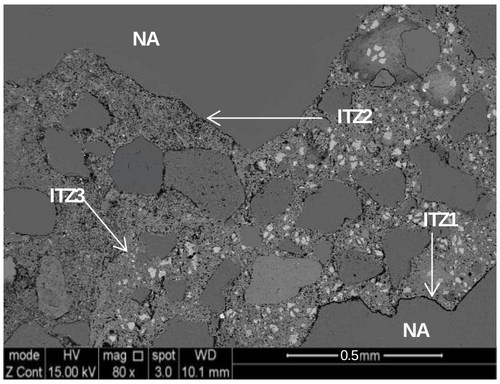


Figure 6: SEM image explains the ITZ s of in RAC sample without Nano silica

On the other hand, the ITZ3 in the RAC specimen (shown in Figure 7) included coated (with SiO_2 nanoparticles) RA particles cannot be clearly identified with a black line. Although, the old attached mortar and the new mortar can be recognized by their different porosity and microstructure, the two parts seems to be connected very well and the zone between them (which is the ITZ3) are very dense and refined (see Figure 6). The densification of the microstructure of the ITZ3 can be attributed to the effect of adding the SiO_2 nanoparticles. These nanoparticles can result in a dense ITZ though two mechanisms: one is physical and the other is chemical. The latter is attributed to the chemical ability of the SiO_2 nanoparticles to react with the $Ca(OH)_2$ (one of the cement hydration products) and leading to the formation of more gel product, calcium silicate hydroxide (C-S-H) which is the product responsible of the strength of the concrete and dense microstructure of the ITZ in concrete (Sanchez & Sobolev, 2010; Said et al., 2012). The former, can be due to large surface area and the super-ultra-fine size of the nanoparticles. This small size of these particles help in filling up all the micro/nano pores and voids exit in the attached mortar and the ITZ resulting in a more densefied microstructure of the cement matrix (Said et al., 2012; Shaikh, Odoh, & Than, 2014). Hence, these two mechanisms seem to be the main reason behind the improvement in the strength of the RAC containing SiO_2 Nanoparticles.



(a)



(b)

Figure 7: a-SEM image of a concrete sample with Nano silica, b- image of the ITZ s of a concrete sample with Nano silica

4. Conclusion

The following conclusions can be drawn based on the results and discussion:

- The addition of Nanoparticles of SiO_2 can improve the compressive strength of recycled aggregate concrete regardless of the content of RA.
- The increase in compressive strength depends on the content of the Nano SiO_2 content.
- The addition of nanoparticles of silica at contents of 0.4%, 0.8% and 1.2% , results in an increase in the compressive strength of 10%, 18% and 20% for mixes containing 50% RA and of 6%, 13% and 16% for mixes made with 100% RA, respectively.
- Microscopic and SEM observations revealed the heterogeneous nature of the RA particles which is characterized by weak and cracked attached mortar and porous ITZ.
- RAC microstructure can be positively modified though the addition of SiO_2 nanoparticles which can help densefy the microstructure RAC by phyiso-chemical mechanisms.
- RAC with Nanoparticles of SiO_2 can demonstrate comparable strength to that of NAC; hence, great enhancement in terms of sustainability and environmental effects of RA can be achieved.

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